

A first group of specifically preferred polyolefins suitable for silane grafting comprises ultra low density polyethylenes (also known as very low density polyethylenes) having a density of less than 0.90 g/cm<sup>3</sup>. Such materials are commercially available, for example, as

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ENGAGE<sup>TM</sup> 8400 (ethylene-co-octene having a density of 0.870 g/cm<sup>3</sup> according to ASTM D-792, a melt flow index (MFI) according to ASTM D-1238 of 30 g/10 min and a melting peak as determined by DSC at a rate of 10°C / min of 60°C);

10 ENGAGE<sup>TM</sup> 8411(ethylene-co-octene having a density of 0.880 g/cm<sup>3</sup> according to ASTM D-792, a melt flow index (MFI) according to ASTM D-1238 of 18 g/10 min and a melting peak as determined by DSC at a rate of 10°C/min of 72°C);

15 ENGAGE<sup>TM</sup> 8401(ethylene-co-octene having a density of 0.885 g/cm<sup>3</sup> according to ASTM D-792, a melt flow index (MFI) according to ASTM D-1238 of 30 g/10 min and a melting peak as determined by DSC at a rate of 10°C/min of 78°C);

20 ENGAGE<sup>TM</sup> 8130(ethylene-co-octene having a density of 0.864 g/cm<sup>3</sup> according to ASTM D-792, a melt flow index (MFI) according to ASTM D-1238 of 13 g/10 min and a melting peak as determined by DSC at a rate of 10°C/min of 50°C). and

ENGAGE<sup>TM</sup> polyolefins are available from Dow DuPont Elastomers (Geneva, Switzerland). Especially preferred materials from this group are ENGAGE<sup>TM</sup> 8400 and ENGAGE<sup>TM</sup> 8407.

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The ultra low density polyethylene polymer is preferably selected so that the melting peak of the polymer as measured by DSC at a rate of 10°C/min is less than 100°C, more preferably less than 90°C and especially preferably between 60°C and 80°C.

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A second group of preferred polyethylenes suitable for silane grafting and specifically preferred for use in the precursor of the present invention comprises copolymers of ethylene and (meth)acrylate monomers, available, for example, as

LOTRYL™ 35 BA 40, a copolymer of ethylene and butyl acrylate (co-E-BA) in a ratio of 65 parts ethylene to 35 parts butyl acrylate having a density of 0.930 g/cm<sup>3</sup> and a melt flow index (MFI) according to ASTM D-1238 (190°C) of 40 g/10 min);

- 5    LOTRYL™ 17 BA 07, a copolymer of ethylene and butyl acrylate (co-E-BA) in a ratio of 83 parts ethylene to 17 parts butyl acrylate having a density of 0.930 g/cm<sup>3</sup> and a melt flow index (MFI) according to ASTM D-1238 (190°C) of 6.5-8 g/10 min);
- 10    LOTRYL™ 28 BA 175, a copolymer of ethylene and butyl acrylate (co-E-BA) in a ratio of 72 parts ethylene to 28 parts butyl acrylate having a density of 0.930 g/cm<sup>3</sup> and a melt flow index (MFI) according to ASTM D-1238 (190°C) of 150-200 g/10 min); and
- 15    LOTRYL™ 28 MA 07, a copolymer of ethylene and methyl acrylate (co-E-MA) in a ratio of 72 parts ethylene to 28 parts methyl acrylate having a density of 0.930 g/cm<sup>3</sup> and a melt flow index (MFI) according to ASTM D-1238 (190°C) of 6-8 g/10 min).
- 20    LOTRYL™ copolymers are available from ATOFINA (Duesseldorf, Germany).

Other suitable ethylene-co-(meth)acrylate polymers suitable for silane grafting and suitable for use in the precursor of the present invention are those which also comprise polymerized units of maleic anhydride. These materials are 25 commercially available from Elf Atochem (Puteaux, France) as

LOTADER™ 6200, a terpolymer of ethylene, acrylic ester and maleic anhydride, having a comonomer content of 9%, a melt flow index of 40 g/10 min and a melting point by DSC of 102°C;

LOTADER™ 8200, a terpolymer of ethylene, acrylic ester and maleic anhydride, having a comonomer content of 9%, a melt flow index of 200 g/10 min and a melting point by DSC of 100°C;

5 LOTADER™ 5500, a terpolymer of ethylene, and acrylic ester and maleic anhydride having a comonomer content of 22%, a melt flow index of 20 g/10 min and a melting point by DSC of 80°C; and

10 LOTADER™ 7500, a terpolymer of ethylene, and acrylic ester and maleic anhydride having a comonomer content of 20%, a melt flow index of 70 g/10 min and a melting point by DSC of 76°C.

15 Preferably a single polymer having a melt flow index of 10-100 g/10 min, such as LOTRYL™ 35 BA 40, is selected from the group of ethylene-co-(meth)acrylate polymers.

The melting point of polymers selected from the group of ethylene-co-(meth)acrylate polymers is preferably less than 100°C and more preferably less than 80°C.

20 The crosslinkable polymers or polymers for use in the precursor of the thermally-conductive material of the present invention are preferably selected to give flexibility to the thermally-formed and crosslinked material. Flexibility is important to allow conformability of the adhesive tape to rough surfaces and to allow it to fill uneven or irregular bonding spaces or gaps. Polymers having a glass transition temperature, T<sub>g</sub>, in the range of between -60°C and -10°C in the 25 uncrosslinked state often exhibit such desirable characteristics.

The polymer is also preferably selected to impart strength to the polymeric film backing so that the resulting thermally-conductive adhesive tape can be handled, converted and applied to the substrates without stretching or breaking.

30 The polymer is also selected to be thermally stable. Preferred polymers for use in the precursor are those that are thermally stable (no weight loss) as measured by thermogravimetric analysis (TGA) at 350°C in air, according to DIN IEC 60811-4-1.

of less than 0.4 W/m-K cannot be relied upon to transmit thermal energy with the efficiency and speed required by the use conditions in the electronics industry.

The invention also provides a thermally-conductive adhesive tape having a thermal impedance of less than 6.0°C-cm<sup>2</sup>/W as measured according to ASTM D

5 5470-95.

Preferred are adhesive tapes which have a dielectric strength of greater than 55 kV/mm, an effective thermal conductivity of at least 0.4 W/m-K and a thermal impedance of less than 6.0°C-cm<sup>2</sup>/W.

The invention also refers to the use of the thermally-conductive crosslinked 10 films for forming a thermally-conductive interface between two substrates, such as the surface of a heat-generating body and a heat-absorbing body, for example. The thermally-conductive crosslinked film which is obtainable by forming the precursor into the shape of a film with subsequent crosslinking, preferably comprises one or more adhesive layers to form an adhesive tape.

15 The invention also refers to a specific use, where the heat-generating body is a printed circuit board and the heat-absorbing body is a heat sink.

The invention also refers to an assembly comprising the thermally-conductive film of the present invention in a bonding relationship between two substrates.

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#### Materials used in the Examples and Comparative Examples

##### A. Crosslinkable Polymers

ENGAGE 8400 - ethylene-co-octene having a density of 0.870 g/cm<sup>3</sup>, available from Dow Dupont Elastomers (Geneva, Switzerland). Melt flow index 25 (MFI) according to ASTM D-1238 of 30 g/10 min.

LOTRYL EA 35 BA 40 - ethylene and butyl acrylate (co-E-BA) in a ratio of 65 parts ethylene to 35 parts butyl acrylate having a density of 0.930 g/cm<sup>3</sup>, available from ATOFINA (Duesseldorf, Germany). Melt flow index (MFI) according to ASTM D-1238 of 40 g/10 min.

30 LOTRYL 28 MA 07 - ethylene and methyl acrylate (co-E-BA) in a ratio of 72 parts ethylene to 28 parts methyl acrylate having a density of 0.95 g/cm<sup>3</sup>,

available from ATOFINA (Duesseldorf, Germany). Melt flow index (MFI) according to ASTM D-1238 of 7 g/10 min.

*B. Vinyl silane of the formula (I)*

5        Vinyl-trimethoxy silane, available as DYNASYLAN VTMO from Degussa AG (Hanau, Germany).

*C. Free-radical initiator*

10      Dicumylperoxide or 2,2-bis-phenylpropyl peroxide, available as Luperox DCSC (active oxygen level of 5.80-5.92 wt. %) from Atofina Deutschland GmbH (Gunzburg, Germany).

*D. Catalysts for moisture-curing (optional)*

15      Tin (II) ethyl hexanoate, available from Johnson Matthey GmbH (Karlsruhe, Germany)

*E. Thermally-conductive filler*

20      MAGNIFIN H5A, Mg(OH)<sub>2</sub> powder, vinyl silane-treated, available from Alusuisse Martinswerk GmbH (Bergheim, Germany). Specific Surface (BET) of 4.0–6.0 m<sup>2</sup>/g. Particle Size: d<sub>50</sub> 1.25–1.65 µm.

Test Methods:

*Breakdown voltage, kV*

25      The breakdown voltage for the adhesive tapes was measured according to DIN (Deutsche Industrie Norm) EN 60243-1. The results were recorded in kV.

*Dielectric strength, kV/mm*

30      The breakdown voltage of adhesive tapes was measured according to DIN EN 60243-1. The results were normalized to account for the thickness of the adhesive tape measured.

Claims

1. Thermally-formable and cross-linkable precursor of a thermally-conductive material comprising

5 a) one or more crosslinkable polymers where the melt flow index of the polymer or mixture of polymers (measured at 190°C according to ASTM D-1238), respectively, is 10-100 g/10 min and  
b) one or more thermally-conductive fillers in an amount of at least 60 wt. % of the total weight of the precursor.

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2. Precursor according to claim 1 wherein the crosslinkable polymers are selected from a group consisting of polyolefins and polyurethanes.

15 3. Precursor according to any of claims 1-2 wherein the crosslinkable polymer is a polyolefin having at least 30 % by weight ethylene units.

4. Precursor according to any of claims 1-3 wherein the polyolefin is selected from the group comprising copolymers of ethylene and (meth)acrylate esters.

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5. Precursor according to any of claims 1-4 wherein at least one of the crosslinkable polymers comprises one or more moisture-curable groups.

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6. Precursor according to claim 5 wherein the moisture-curable groups comprise silane groups.

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7. Precursor according to claims 5-6 wherein the crosslinkable polymer is obtainable by reacting a polymer according to any one of the claims 1-6 with one or more vinyl silane compounds of the formula  $RR'SiY_2$  (I), wherein R is a monovalently olefinically unsaturated radical, R' is a monovalent radical free of aliphatic unsaturation and Y is a hydrolyzable organic radical, and a free-radical initiator.

8. Precursor according to claim 7 wherein the vinyl silane compound(s) (I) are employed in an amount of at least 2 parts per 100 parts crosslinkable polymer or polymers.

5 9. Precursor according to any of the claims 7-8 wherein the free-radical initiator is selected from the group consisting of an organic peroxide and an organic perester.

10 10. Precursor according to claim 9 wherein the free-radical initiator is employed in the amount of at least 0.1 parts per 100 parts crosslinkable polymer or polymers.

15 11. Precursor according to any of claims 5-10 comprising a catalyst for moisture-curing of the moisture-curable group in an amount of greater than 0.05 wt. % based on the total weight of the precursor.

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12. Precursor according to any one of claims 1-11 wherein the thermally-conductive filler is selected from a group consisting of alumina, aluminum oxide, aluminum trihydroxide and magnesium hydroxide.

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13. Method of manufacturing the precursor of any of the claims 1-12 comprising the steps of:

- providing one or more crosslinkable polymers where the melt flow index of the polymer or mixture of polymers (measured at 190°C according to ASTM D-1238 ), respectively, is 10-100 g/10 min and
- compounding the polymer or polymers with one or more thermally-conductive fillers in an amount of 60 wt. % of the total weight of the precursor in a heated mixing device.

14. Method of manufacturing the precursor of any of the claims 7-12 comprising the steps of:

- a) providing one or more crosslinkable polymers where the melt flow index of the polymer or mixture of polymers (measured at 190°C according to 5 ASTM D-1238 ), respectively, is 10-100 g/10 min and where at least one of the polymers has an ethylene unit content of at least 30 % by weight,
- b) reacting the polymer with a vinyl silane of the formula (I), wherein R is a monovalently olefinically unsaturated radical, R' is a monovalent radical 10 free of aliphatic unsaturation and Y is a hydrolyzable organic radical, and a free-radical initiator in a heated mixing device to produce a moisture-curable polymer and
- c) compounding the moisture-curable polymer or polymers, respectively, with one or more thermally-conductive fillers in the amount of at least 15 60 wt. % in a heated mixing device.

15. Method of manufacturing a shaped thermally-conductive material comprising the steps of:

- a) providing the precursor of any of the claims 1-12,
- b) thermally forming the precursor to a desired shape and
- c) crosslinking the precursor.

16. Method according to claim 15 wherein cross-linking is effected by applying  $\gamma$ -irradiation.

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17. Method according to claim 16 wherein the  $\gamma$ -irradiation has an energy of between 50 keV-25 MeV.

30 18. Method according to any of claims 16-17 wherein the  $\gamma$ -irradiation is applied to the precursor in a dosage of at least 50 kGy.

19. Method according to claim 15 wherein cross-linking is effected by moisture-curing.
20. Method of any of claims 15-19 where the precursor is thermally-formed into  
5 the shape of a film by extrusion.
21. Thermally-conductive material obtainable by the method of any of the claims  
13-20.
- 10 22. Adhesive tape comprising at least a film backing bearing an adhesive layer  
on at least one of the major surfaces of the film backing, wherein the film  
backing is obtainable by extruding the precursor according to any of the  
claims 1-12 into the shape of a film and crosslinking the film.
- 15 23. Adhesive tape according to claim 22 having a dielectric strength of at least  
55 kV/mm as measured according to DIN EN 60243-1.
24. Adhesive tape according to any of the claims 21-23 having an effective  
thermal conductivity of at least 0.4 W/m-K as measured according to ASTM  
20 D 5470-95.
25. Adhesive tape according to any of the claims 21-24 having a thickness of  
less than 300  $\mu\text{m}$ .
- 25 26. Adhesive tape according to any of the claims 21-25 where the crosslinked  
thermally-conductive material of the film backing has an elastic torque,  $S'$ , of  
between 5 dNm and 8 dNm, as measured according to ASTM D 6294-9.
27. Adhesive tape of any of the claims 21-26, wherein the adhesive is a  
30 pressure-sensitive adhesive.

28. Use of an adhesive tape according to any of the claims 21-27 for providing thermal conductivity between two substrates.
29. Assembly comprising the adhesive tape according to any of the claims 21-27  
5 in a bonding relationship between two substrates.